EOS MLS Observations of ozone loss in the 2004-2005 Arctic Winter

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Abstract. Earth Observing System Microwave Limb Sounder O_3 and N_2O are used to examine transport and chemical O_3 loss in the unusually cold 2004-2005 Arctic winter. The vortex was dynamically active, with episodic mixing events throughout the winter; descent was the dominant transport process only through late January. Before the onset of lower stratospheric chemical loss, O_3 was higher near the vortex edge than in the vortex core, causing different effects of mixing depending on the vortex region and time, either masking or mimicking chemical loss. O_3 loss ceased by 10 March because of an early final warming. Rough estimates suggest maximum vortex-averaged O_3 loss of 1.2-1.5 ppmv between 450 and 500 K, with up to $\sim$2 ppmv loss in the outer vortex near 500 K. Despite record cold, chemical O_3 loss was less in 2004-2005 than in previous cold Arctic winters.

1. Introduction

The 2004-2005 Arctic winter lower stratosphere was the coldest on record, with potential for polar stratospheric cloud (PSC) formation on more days and over a larger region than in any previously observed northern hemisphere (NH) winter (Figure 1). It was the first NH winter observed by the Earth Observing System (EOS) Microwave Limb Sounder (MLS). EOS MLS is a successor to the Upper Atmosphere Research Satellite (UARS) MLS, with greatly enhanced capabilities for studying polar O_3 [*Waters et al.*, 2005]: O_3 is measured with greater precision and temporal and spatial coverage than UARS MLS, without gaps in the polar regions due to satellite yaws; EOS MLS N_2O measurements provide a long-lived tracer for diagnosing transport, critical to assessing Arctic O_3 loss [e.g., *WMO*, 2003].

EOS MLS observations indicate extensive PSC activity after mid-December 2004 [*Santee, et al., "A study of stratospheric chlorine partitioning in the winter polar vortices based on new satellite measurements and modeling", in preparation, hereinafter SA*], including evidence of ice PSCs in late January. Studies of chlorine from MLS and ACE-FTS (the Atmospheric Chemistry Experiment-Fourier Transform Spectrometer) [*Dufour et al, "Partitioning between the inor-*
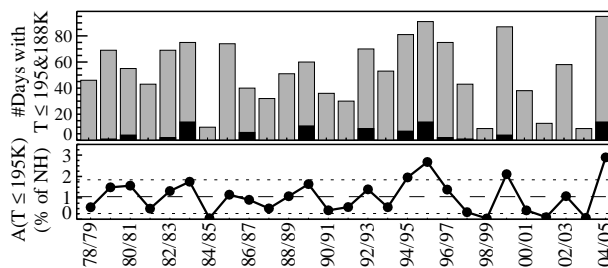

Figure 1. (Top) Number of days with temperature less than the threshold for nitric acid trihydrate (NAT, light bars) and ice (dark bars) PSCs at 50 hPa, and (bottom) area averaged over December through March where NAT PSCs could form at 50 hPa, for 1978-1979 through 2004-2005; thin dashed and dotted lines show average over the years and one-standard deviation envelope, respectively. From National Centers for Environmental Prediction/Climate Prediction Center data.

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ganic chlorine reservoirs HCl and ClONO$_2$ during the Arctic winter 2005 derived from ACE-FTS measurements", submitted to Atmos. Chem. Phys.; SA] indicate strong chlorine activation from early January through early March 2005, after which low temperatures and PSC existence were halted by a "major final warming" (MFW, a major warming that progresses directly into the final warming [e.g., *Labitzke and Collaborators*, 2002]). Here we discuss evidence for chemical O$_3$ loss using MLS O$_3$ and N$_2$O observations viewed in relation to the vortex evolution.

The MLS data are the first publicly released version (v1.51). In the lower stratosphere, vertical resolution is $\sim$3-4 km for N$_2$O and O$_3$, and estimated precisions are $\sim$20-30 ppbv and $\sim$0.2-0.3 ppmv, respectively; early validation results show O$_3$ agrees to within 5-15% with other datasets, and N$_2$O agrees with balloon-borne observations within the expected precisions [*Froidevaux et al.*, 2005]. For maps and time series, MLS data are gridded at 2×5° (latitude×longitude) using a weighted average around each gridpoint of 24 hours' data. NASA's Global Modeling and Assimilation Office Goddard Earth Observing System Version 4.0.3 (GEOS-4) meteorological analyses [*Bloom et al.*, 2005] are used for potential vorticity (PV), equivalent latitude (EqL, the latitude that would enclose the same area between it and the pole as a given PV contour) mapping, and interpolation to isentropic (potential temperature, θ) surfaces. A few days when MLS data are unavailable are filled using a Kalman filter and are indicated by pale colors in the time series plots [e.g., *Santee et al.*, 2003].

2. MLS Observations of Vortex Evolution and Ozone Loss

Figure 2 shows 490 K ($\sim$19 km) MLS N$_2$O and O$_3$ maps during the 2004-2005 winter. The time evolution of N$_2$O and O$_3$ is shown in more detail in Figure 3. Chlorine was not substantially activated until late December 2004 [*SA*]. Prior to significant chlorine activation, a region of lower O$_3$ developed in the vortex core (via transport processes under investigation) during vortex formation. The vortex was frequently dynamically active during this winter. In early to mid-January and throughout February, evidence of mixing was seen in excursions of higher N$_2$O deeper into the vortex (Figure 3). This is reflected in the 29 January maps, which suggest intrusions of extravortex air into the outer vortex (near 0° longitude) and intrusions of vortex edge air into the core (near 30°E). N$_2$O began to increase throughout the vortex after about 1 February, indicating that descent was no longer the dominant transport process. Figure 3 shows excursions of high N$_2$O into the vortex core starting in late February (also seen in 27 February map, near 140 and
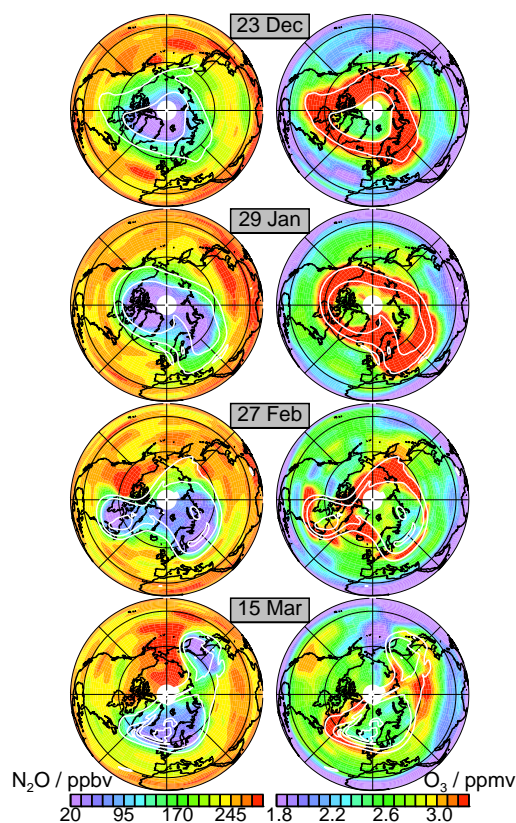

Figure 2. 490 K MLS N$_2$O and O$_3$ maps in 2004-2005. White contours are PV near the outside and inside of the region of strong gradients demarking the vortex edge.
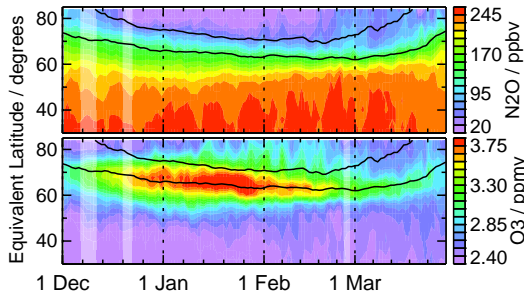
Figure 3. EqL-time plots of 490 K MLS N_2O and O_3 during the 2004-2005 NH winter. Overlaid contours are scaled PV (sPV, *Manney et al.* [1994]), located inside the vortex edge and near the edge of the vortex "core", as defined below for Figures 4 and 6.

340°E), with the vortex shrinking rapidly after late February and becoming even more distorted and variable in early March. By 15 March, with the MFW in progress, MLS observations showed that chlorine had been deactivated [*SA*]; the lower stratospheric vortex was split, and large tongues of midlatitude air extended into the vortex region.

Between 23 December and 29 January, N_2O (O_3) decreased (increased) in the vortex, indicating that diabatic descent was the dominant process during this period. Figure 3 shows that O_3 began decreasing in mid-January and N_2O increased after early February. Since N_2O was still decreasing due to descent in mid-January, the O_3 decrease starting at this time is attributed to the onset of chemical O_3 loss. O_3 continued to decrease through ~10 March, with the rate of decrease accelerating (especially in the vortex core) after late February. The increase in vortex N_2O starting in February (apparent in 27 February map) indicates the increasing importance of transport processes other than descent. Some episodic decreases in O_3 in the outer part of the vortex (e.g., late February) are coincident with increases in vortex N_2O, indicating intrusions; in January and early February, temporary increases of O_3 in the vortex core are associated with increases in N_2O resulting from intrusions of vortex edge air into the core (e.g., 29 January). Figure 4 summarizes the average evolution of the vertical distributions of N_2O and O_3 in the vortex. The "vortex" average shows the entire region within the equatorward PV contour in Figure 3, whereas the "outer vortex" average shows the region between those two PV contours. Similar results were obtained using averages within EqL bands (≥62°EqL for vortex, 62-72°EqL for outer vortex), but these bands do not isolate a consistent portion of the vortex throughout the winter (especially in early and late winter, Figure 3). The descending N_2O contours confirm that descent was the dominant process affecting N_2O below ~20 km through late January in both regions; at higher levels, effects of mixing were seen

in the vortex edge region through most of the winter. The mixing events in early February and early March resulted in strong N_2O increases at the lowest levels in both regions; rapid increases after early March reflect the beginning of the vortex breakup. O_3 decreased fairly monotonically starting in mid-January, before N_2O began to increase; O_3 decreased slightly more in the outer vortex than in the vortex average, especially above ~20 km. The reason for the O_3 decrease in early February near 550-650 K in the outer vortex is unclear; while O_3 was on average higher along the vortex edge at these levels, it was not uniformly so, thus mixing cannot be ruled out. Also, since active chlorine extended above 600 K at times [*SA*], chemical loss may have occurred.

Figure 5 summarizes the net observed changes over the period of chemical O_3 loss. Descent was dominant (N_2O decreased) in the lower stratosphere below ~600 K (~25 km) only in the outer part of the vortex; in the vortex core, there was a net increase in N_2O due to mixing of air from further towards the vortex edge. At higher levels, mixing dominated (N_2O increased) along the vortex edge, with descent dominating in the vortex core above ~26 km. Since descent increases with altitude, less mixing is needed to compensate for it at lower levels. O_3 decreased throughout the vortex below about 500 K, and along the vortex edge up to ~600 K. The decrease in the outer vortex region was in the area where descent (which increases O_3) dominated, and thus can be attributed to chemical loss, which was partially masked by transport. Since O_3 in the vortex core was initially lower than in the outer vortex, both mixing and descent tended to increase O_3 there; thus the decrease in the core again indicates chemical loss that may have been partially masked by transport processes. The maximum observed O_3 decrease was ~1.2 ppmv in the outer vortex near 500 K. The complexity of the transport processes apparent in N_2O, coupled with the morphology of O_3 (which caused mixing to produce changes of different signs in different portions of the vortex), makes distinguishing between transport processes and chemical loss even more challenging than during more typical NH winters.

Figure 6 shows 23 January and 10 March profiles from the averages in Figure 4. Maximum observed O_3 decreases in the outer vortex average were ~1.1 ppmv near 500 K; in the vortex average, maximum decreases were only slightly smaller, nearly 1.0 ppmv, but at a lower level, near 450 K. If we assume, as is commonly done, that vortex N_2O changed primarily as a result of descent, then the descent rate estimated from those N_2O changes can be applied to the O_3 profile to estimate chemical loss. Such estimates suggest up to 2 ppmv chemical loss in the outer vortex, and ~1.5 ppmv loss in the vortex average, with maximum losses in both regions between 450 and 500 K. We have already shown, how-
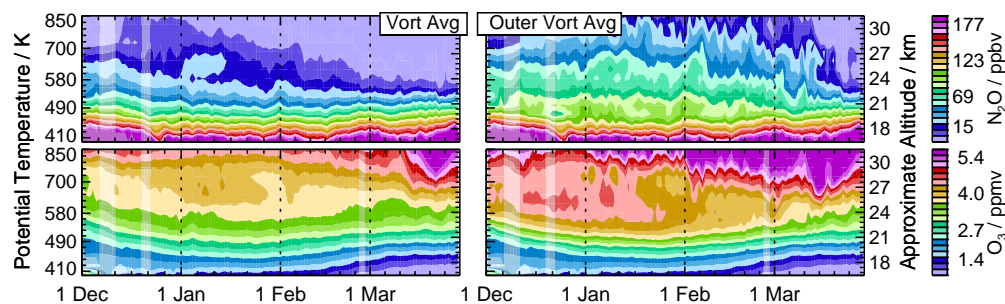
Figure 4. Timeseries of average MLS N_2O and O_3 for sPV $\geq 1.6 \times 10^{-4}s^{-1}$ (throughout vortex) and for $1.6 \leq sPV \leq 2.2 \times 10^{-4}s^{-1}$ (in outer ring of vortex).
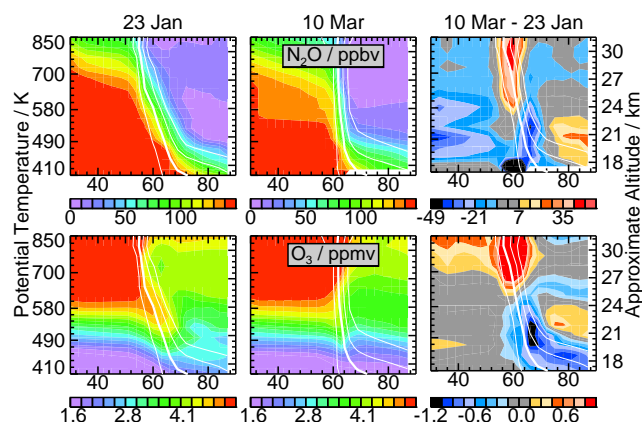

Figure 5. EqL/θ cross-sections of MLS N_2O and O_3 during the 2004-2005 NH winter. Overlaid contours are sPV; thick contour shows approximate vortex edge location, coincident with outer contour in Figure 3. Outer vortex average in Figure 4 is in region between thick and innermost PV contours.

Figure 6. Average MLS N_2O and O_3 profiles in regions given in Figure 4 on 23 January 2005 (red) and 10 March 2005 (black) for vortex (solid) and outer vortex (dot-dashed, offset by +50 ppbv/+1 ppmv for N_2O/O_3).

ever, that mixing substantially altered N_2O this winter, so these estimates are more uncertain than those for winters in which descent primarily controlled N_2O changes. The outer vortex N_2O profiles between about 460 and 520 K, as well as Figure 5, do suggest that descent was dominant there, so the estimated maximum loss of $\sim$2 ppmv in this region may be more robust.

O_3 changes were also examined as a function of N_2O, indicating local O_3 decreases up to 2-2.5 ppmv at 100 to 120 ppbv N_2O; these changes cannot be confidently attributed to chemical loss since mixing processes can cause similar changes in O_3/N_2O correlations [e.g., *Michelsen et al.*, 1998].

A slightly different method is used to compare O_3 loss estimates for this winter with those from the Polar Ozone and Aerosol Measurement (POAM) III solar occultation instrument and from previous years. As described by *Hop-*
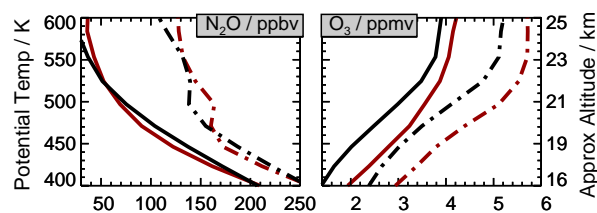
pel et al. [2002], vortex-averaged descent is estimated using three-dimensional trajectory calculations and descent rates from a radiation code; the difference between an initial profile descended with these rates and a final observed profile gives an estimate of chemical loss. Figure 7 compares estimates from EOS MLS with those from POAM II in 1996, POAM III in 2000 and 2005, and UARS MLS in 1996. 1996 and 2000 were the previous years with the most observed Arctic O_3 loss [e.g., *WMO*, 2003]. The only difference between the POAM and MLS calculations is the initial and final profiles used. Since POAM observes only a narrow latitude band each day, the representativeness of such a "vortex-average" profile depends on the meteorological conditions and O_3 morphology. In 2005, POAM and MLS results agree well only below $\sim$470 K, possibly suggesting that POAM sampling was less representative of the vortex at higher altitudes this winter. The close agreement in 1996 suggests that POAM sampling was representative of the whole vortex then, when the vortex was less variable and O_3 within it more uniform [*Manney et al.*, 2003]; estimates for 1996 may be less sensitive to sampling than those for 2005.

The 2005 estimates suggest maximum vortex-averaged chemical O_3 loss near 450 K of 1.2-1.3 ppmv. This is slightly less than the estimates from MLS N_2O and O_3 above, suggesting that the calculations using trajectories and the radiation code may underestimate descent. The maxi-
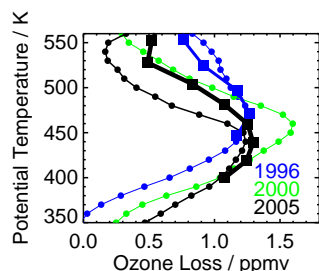
Figure 7. Chemical O$_3$ loss from 5 January through 10 March estimated using "vortex-averaged descent" (see text) from POAM (thin lines with dots) for 1996, 2000, and 2005, and from UARS MLS for 1996 (for 25 December 1995 through 3 March 1996) and EOS MLS for 2005 (thick lines with squares).

mum loss from UARS MLS in 1996 from this method is also slightly less than the $\sim$1.4 ppmv found by *Manney et al.* [2003] for the same period, but within the uncertainties; these estimates agree well with those from vortex averages of ozonesonde data for 1996 and 2000 ($\sim$1.2 and 1.5 ppmv, respectively, by early March), and find slightly lower loss than the estimate from Match for 2000 (about 2.0 ppmv by 10 March) [*WMO*, 2003]. The maximum chemical loss estimated here for 2005 is comparable to that in 1996, but largest loss in 2005 was at lower altitudes; considerably more loss (up to $\sim$1.6 ppmv near 460 K) was seen in 2000. In addition, in both 1996 and 2000, O$_3$ loss continued after 10 March [e.g., *WMO*, 2003], whereas in 2005 this represents the complete period of O$_3$ loss. Thus, despite lower temperatures, the combination of conditions in 2005 (vortex structure and evolution, and early cessation of processing) resulted in less chemical O$_3$ loss than in the coldest previous NH winters.

3. Summary

EOS MLS observations of O$_3$ and N$_2$O during the unusually cold 2004-2005 winter provide a detailed view of transport and chemical O$_3$ loss throughout the winter. Although the winter was the coldest on record, mixing processes were more important in determining trace gas transport in the vortex than during previous cold Arctic winters. MLS N$_2$O observations show mixing events throughout the winter; descent was the dominant transport process only until late January. Prior to the onset of chemical loss, O$_3$ was higher near the lower stratospheric vortex edge than in the vortex core; as a result, mixing processes either masked or mimicked chemical loss, depending on the part of the vortex in which they occurred.

Polar processing ceased by 10 March because of an early final warming. Rough estimates of chemical loss using N$_2$O observations and vortex-averaged descent rates to estimate

changes due to transport suggest maximum vortex-averaged O$_3$ loss of 1.2-1.5 ppmv between 450 and 500 K. In the outer vortex where observed decreases were largest and where descent dominated N$_2$O changes throughout the winter, MLS data suggest localized chemical loss up to $\sim$2 ppmv. Estimates using a vortex-averaged descent method for POAM and MLS data imply that O$_3$ loss in 2004-2005 was comparable to that in 1996 (but with the maximum loss at lower altitude) and less than that in 2000, the two previous years with most Arctic O$_3$ loss. Thus, initial expectations (and reports in the popular press) of unprecedented O$_3$ loss during this coldest winter did not, in fact, materialize because of dynamical factors, including enhanced mixing and an early final warming. Because of the complexity of the transport processes and the O$_3$ morphology, a more precise determination of chemical loss during the 2004-2005 Arctic winter will require extensive modeling and data analysis efforts.

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